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A Study of the Particulate and Gaseous Emissions of Tritium from Neutron Generator Targets

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In the present study neutron generator targets were examined to determine the extent of particulate and gaseous emission of tritium. To determine whether deuteron bombardment of the targets may affect the tritium emissions, both new and used targets were studied.

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A Study of the Particulate and Gaseous Emissions of Tritium from Neutron Generator Targets

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Introduction

NEUTRON generator targets are prepared by adsorption of elemental tritium on titanium or zirconium film on a copper backing. The stability of tritium on the target depends on the ability of Ti or Zr to form stable hydrides (tritides) resulting in chemical compounds rather than a mere physical adsorption of the gas. This chemical stability presumably is also the cause of the tritium being released in particulate form.

In the present study neutron generator targets were examined to determine the extent of particulate and gaseous emission of tritium. To determine whether deuteron bombardment of the targets may affect the tritium emission, both new and used targets were studied.

Procedures and Methods

The targets used in this study were all obtained from Texas Nuclear Corporation and consisted of tritium adsorbed on titanium with a copper backing, 0.4–0.8 Ci/cm^2 on an area of approx 5 cm^2. Four of the targets had been exposed to deuteron bombardments ranging from 12 to 220 mA-min.

The experimental setup for the study of the tritium emission is shown in Fig. 1. Air is drawn through flowmeter M at 2 l/min and subsequently through microfilter F-1 to remove airborne particles. The filtered air then flows over the target, contained in target holder H, and then through microfilter F-2 to trap particles emitted by the target. The air then flows through C, a heated tube approx 70 cm long ×2.6 cm O.D., packed with 125 G of a catalyst, consisting of 0.5% platinum on 0.32 cm aluminum pellets (obtained from Engelhard Industries of Newark, N.J.). In the presence of oxygen and at temperatures above 200°C the catalyst oxidizes hydrogen gas to water. The air then passes through microfilter F-3 to remove any particles originating from the catalyst and into trap A-1 which is held at liquid nitrogen temperatures to freeze out the water. The air pump completes the train. The microfilters had a diameter of 47 mm and a pore size of 0.2 μm. They were obtained from Gelman Instrument Co., Ann Arbor, MI (Metricel GA-8).

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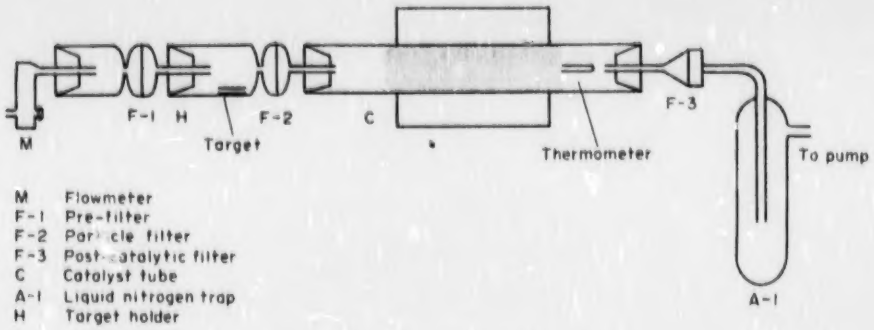

FIG. 1. Emission separation train.

In order to trap the particles on the glass the target holder was coated with Apiezon-N grease by dipping the target holder into a beaker containing a solution of 1 g of the grease in 500 ml of a toluene mixture. Evaporation of the solvent on slow removal of the target holder from the solution left a coating of Apiezon-N on the glass.

Usually air was allowed to flow through the train for 4 hr to accumulate sufficient activity for accurate measurement. Subsequently, microfilters F-1, F-2, and F-3 were removed and placed into liquid scintillation counting vials. To each vial was added 20 ml of a dioxane based scintillation liquid (Mo69). The grease coating in the target holder was dissolved in ten 10-ml washings of toluene, each washing being caught in a different counting vial. To each of these washings was added 10 ml of the counting solution. Water from the liquid nitrogen trap was weighed and dissolved in enough of the dioxane based scintillation liquid to yield 20 ml of solution. If more than 5 ml of water was obtained, aliquots were taken for counting. An internal standard method with tritiated hexadecane was used for the determination of counting efficiency. To establish possible cross-contamination a blank run was carried out between any two target runs. The blank consisted of an overnight run during which no target was present in the target holder.

During two of the runs an additional liquid nitrogen trap was inserted between microfilter F-2 and the catalyst in an attempt to capture any tritium emitted from the target in the form of water. When used, the trap was treated in the same way as A-1 but labled A-2.

Results and Discussion

Table 1 gives the tritium activities from the various targets studied with the averages of the activities found over several runs on the same targets. Filters F-1 and F-3 should contain no activity above the expected radon daughters from air and, in general, this activity was low when compared with the activity found in other parts of the train. Any activity above background found on these two filters was probably representative of some cross-contamination in spite of the rigid cleaning operations performed on all glassware between runs. The blanks run between target samples showed activities of approximately the same orders of magnitude as those shown in Table 1 for filters F-1 and F-3, higher blanks being observed in all cases after a run with a used target.

In analyzing the data, it is assumed that the total particulate emission is represented by the sum of the activities on filter F-2 and the washes of the target holder. The total gaseous emission was taken to be the sum of the activities in traps A-1 and A-2, where the latter was used.

In certain cases tritium activity was too high for measurement by liquid scintillation counting or for efficiency determination by the internal standard method. These samples were diluted and an aliquot was taken for measurement. This technique is inherently inaccurate for measurement of activity of tritium in particulate form, particularly, if the entire activity is associated with a few particles.

Experimentally this is observed during the dilution of the sample. An aliquot contains an unreasonably large or small counting rate. Thus, counting is done for a few seconds and a typical counting efficiency is estimated. However, it is estimated that the reported values are accurate to within a factor of two. The inaccuracies are probably on the high side because the quenching of the sample in the liquid scintillation counter could be higher than estimated whereas the upper limit of the counting efficiency is given by the inherent limitations of the system.

Table 1. Tritium emission from targets in dis/min

Target	Filter F-1	Filter F-2	Filter F-3	Water A-1	Water A-2	Totals of Washes of H
T-18	5.4×10^2	2.05×10^3	BKG	1.5×10^4		2.0×10^3
(Used)	1.2×10^2	2.9×10^3	BKG	7.9×10^4		4.7×10^6
	1.9×10^3	$7.4 \times 10^{4*}$	BKG	4.4×10^4		
AVERAGES	0.8×10^2	2.6×10^4	BKG	4.6×10^4		2.4×10^5
T-19	6.5×10^2	$3.5 \times 10^{8*}$	7.8×10^2	3.8×10^5	3.5×10^7	$1.7 \times 10^{7*}$
(Used)	BKG	$2.6 \times 10^{8*}$	4.9×10^2	$1.9 \times 10^{7*}$	$1.1 \times 10^{8*}$	$1.7 \times 10^{8*}$
AVERAGES	3.2×10^2	3.0×10^8	6.4×10^2	3.8×10^5	7.2×10^7	9.4×10^7
T-21	BKG	$3.9 \times 10^{8*}$	1.2×10^3	3.2×10^6		
(Used)	BKG	$1.3 \times 10^{8*}$	4.0×10^2	1.5×10^7		$1.8 \times 10^{8*}$
	1.4×10^2	$8.5 \times 10^{8*}$	1.6×10^4	3.3×10^7		$1.5 \times 10^{8*}$
AVERAGES	BKG	4.6×10^8	7.1×10^3	1.7×10^7		1.6×10^8
T-22	BKG	$2.6 \times 10^{7*}$	2.0×10^2	$9.3 \times 10^{6*}$	$1.4 \times 10^{7*}$	$6.1 \times 10^{7*}$
(Used)	7.4×10^3	$1.6 \times 10^{7*}$	BKG	$3.3 \times 10^{5*}$	$7.9 \times 10^{6*}$	$3.2 \times 10^{7*}$
AVERAGES	3.7×10^3	2.1×10^7	BKG	4.8×10^6	1.1×10^7	1.9×10^8
T-25	BKG	2.8×10^2	BKG	2.2×10^4		
(New)	2.9×10^4	1.3×10^3	BKG	0.7×10^4		5.0×10^4
	1.4×10^3	1.5×10^3	3.6×10^3	6.7×10^4		2.6×10^4
AVERAGES	1.0×10^4	1.0×10^3	1.2×10^3	5.9×10^4		3.8×10^4
T-26	8.9×10^2	1.3×10^3	BKG	1.6×10^3		
(New)	2.5×10^2	1.4×10^3	BKG	8.9×10^4		1.0×10^6
	3.6×1 ?	1.6×10^3	BKG	4.6×10^4		2.1×10^4
AVERAGES	5.0×1 ?	1.4×10^3	BKG	4.6×10^4		1.6×10^4

* Estimated Activity

The hypothesis that such inexact dilutions might be due to the presence of particles is supported by the fact that such estimation was necessary only in runs involving used targets which, as seen later, were found to exhibit higher particulate emission than the new targets.

Table 2 summarizes the results of Table 1 and gives the ratio of particulate to gaseous emissions (*P/G*). The table also shows the total usage of the used targets expressed in units of mA-min. Omitting target T-18 as a possible outlier, the used targets show a gaseous emission of approx 10^3 higher than that of the unused targets. Also, for the used targets a large part of the tritium emission is particulate, rather than gaseous; while for the new targets most of the emission is gaseous. It is difficult to understand the lesser emission of T-18 which has had more use than the other targets, and yet showed less emission. Specific manufacturing conditions could be the reason for the small emission of this target, notably in gaseous form. It is, however, interesting to note that particulate to gaseous ration of tritium emission is high for this target.

Because of the small number of targets studied and because the amount of use of the used targets differed by only an order of magnitude, no reasonable conclusion on differences in emission from targets of different ages could be drawn from the present study.

Table 2. Ratios of particulate to gaseous emissions for the targets studied and total gaseous emissions

Target	Particulate(dpm)	Gaseous Emission(dpm)	Ratio(P/G)	mA-min
T-18	2.4×10^6	4.6×10^4	52	220
T-19	3.9×10^8	7.2×10^7	5.4	110
T-21	0.2×10^8	1.7×10^7	48	12
T-22	2.1×10^8	1.6×10^7	13	16
T-25	3.9×10^4	5.9×10^4	0.66	New
T-26	1.7×10^4	4.6×10^4	0.37	New

Reference

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